

Application No. 10/741,324
Amendment dated: November 29, 2006
Reply to Office Action of August 29, 2006

RECEIVED
CENTRAL FAX CENTER
NOV 29 2006

129420-2

Amendments to the Claims

This listing of claims will replace all prior versions, and listings of claims in the application.

Listing of Claims:

1. (Currently Amended) A method for the preparation of 4,4'-diisopropylbiphenyl, said method comprising:
 - (a) preparing an acidic zeolite catalyst having a molar ratio of SiO₂ to Al₂O₃ in a range between about 20 to 1 and about 500 to 1, and determining an acidic site distribution for said catalyst comprising acidic sites, said distribution comprising 35% or more of said sites having an activation energy of ammonia desorption in a range between about 145 kJ/mol and about 170 kJ/mol;
 - (b) continuously contacting in a flow reactor (i) biphenyl, (ii) at least one inert solvent, (iii) propene, and (iv) an inert diluent gas, said flow reactor containing said acidic zeolite catalyst, said contacting being conducted at a pressure greater than about 1 atmosphere and at a temperature greater than 180°C; and
 - (c) continuously recovering an effluent steam comprising product 4,4'-diisopropylbiphenyl, inert solvent, and inert diluent gas.
2. (Original) The method according to claim 1 wherein said acidic zeolite catalyst is prepared from a precursor zeolite by a method comprising a dealumination step.
3. (Original) The method according to claim 2 wherein, said dealumination step comprises steam treatment.
4. (Original) The method according to claim 3 wherein, said steam treatment is carried out for a duration of at least about one hour at a temperature in a range between about 550°C and about 800°C.
5. (Original) The method according to claim 2 wherein, said dealumination step comprises thermal treatment in the absence of exogenous steam.
6. (Original) The method according to claim 5 wherein, said thermal treatment is carried out for a duration of at least about two hours at a temperature in a range between about 550°C and about 800°C.

Application No. 10/741,324
Amendment dated: November 29, 2006
Reply to Office Action of August 29, 2006

129420-2

7. (Original) The method according to claim 2 wherein said dealumination step comprises contacting with aqueous acid.

8. (Original) The method according to claim 7 wherein, said aqueous acid is selected independently from a group consisting of aqueous sulfuric acid, aqueous nitric acid, aqueous oxalic acid, aqueous sulfuric acid-ammonium nitrate mixtures, and mixtures thereof.

9. (Original) The method according to claim 7 wherein, said contacting with said acid is carried out for a time period in the range of about 0.25 hours to about 4 hours.

10. (Original) The method according to claim 2 wherein, said precursor zeolite is selected from the group consisting of Mordenite, Mazzite, and Beta zeolites.

11. (Original) The method according to claim 10 wherein, said precursor zeolite is a Mordenite zeolite.

12. (Original) The method according to claim 1 wherein, the biphenyl is continuously contacted with the acidic zeolite catalyst at a weight hourly space velocity (WHSV) of between about 0.025 hr^{-1} and about 10 hr^{-1} .

13. (Original) The method according to claim 12 herein said weight hourly space velocity (WHSV) is in a range between about 0.1 and about 2.5 hr^{-1} .

14. (Original) The method according to claim 1 wherein, said inert solvent has a boiling point in a range between about 180°C and about 320°C .

15. (Original) The method according to claim 14 wherein, said inert solvent is a saturated hydrocarbon.

16. (Original) The method of claim 15 wherein, said inert solvent is decalin.

17. (Original) The method according to claim 1 wherein, said propene is present in an amount corresponding to between about 0.1 mole and about 10 moles of propene per mole of biphenyl.

Application No. 10/741,324
Amendment dated: November 29, 2006
Reply to Office Action of August 29, 2006

129420-2

18. (Original) The method according to claim 17 wherein, said propene is present in an amount corresponding to between about 2 and about 5 moles of propene per mole of biphenyl.
19. (Original) The method according to claim 1 wherein, said inert diluent gas is selected from the group consisting of nitrogen, helium, argon, and carbon dioxide.
20. (Original) The method according to claim 1 wherein, said inert diluent gas is nitrogen.
21. (Original) The method according to claim 1 wherein, said diluent gas is present in an amount corresponding to between about 1 mole and about 100 moles of diluent gas per mole of biphenyl.
22. (Original) The method according to claim 1 wherein, said diluent gas is present in an amount corresponding to between about 5 mole and about 100 moles of diluent gas per mole of biphenyl.
23. (Original) The method according to claim 1 wherein, said contacting is conducted at a pressure in a range between about 1 atmosphere and about 100 atmospheres.
24. (Original) The method according to claim 23 wherein, said contacting is carried out at a pressure in a range between about 1 atmospheres and about 40 atmospheres.
25. (Original) The method according to claim 1 wherein, said contacting is carried out at a temperature in a range between about 150°C and about 320°C.
26. (Original) The method according to claim 25 wherein, said contacting is carried out at a temperature in a range between about 200°C and about 250°C.
27. (Original) The method according to claim 1 wherein, said biphenyl and said inert solvent are continuously contacted with said acidic zeolite catalyst at a feed rate corresponding to between about 0.1 and about 5 catalyst bed volumes per hour.

Application No. 10/741,324
Amendment dated: November 29, 2006
Reply to Office Action of August 29, 2006

129420-2

28. (Original) The method according to claim 27 wherein, said feed rate corresponds to between about 1 and about 3 catalyst bed volumes per hour.
29. (Original) The method according to claim 1 having a selectivity for 4,4'-diisopropylbiphenyl of at least 80 percent.
30. (Original) The method according to claim 1 having a yield of 4,4'-diisopropylbiphenyl of at least 50 percent.
31. (Original) The method according to claim 1 wherein said acidic zeolite catalyst is prepared by dealumination of a precursor zeolite, said dealumination comprising the steps of calcinations and treatment with aqueous acid.
32. (Original) The method according to claim 1 wherein said preparing an acidic zeolite catalyst comprises contacting an acidic zeolite with at least one volatile basic agent at temperature range between about 0°C and about 500°C and at a pressure in a range between about 0.1 torr and about 10 atmospheres.
33. (Original) The method according to claim 32 wherein, said volatile basic agent is selected from the group consisting of amines and phosphines.
34. (Original) The method according to claim 33 wherein said amines are selected from the group consisting of ammonia, alkyl amines, dialkyl amines, trialkyl amines, cycloaliphatic amines, aromatic amines, nitrogen-containing heterocyclic aromatic compounds, and nitrogen-containing alicyclic compounds.
35. (Original) The method according to claim 33 wherein said volatile basic agent is ammonia.
36. (Original) The method according to claim 33 wherein said phosphines are selected from the group consisting of phosphine (PH_3), alkyl phosphines, dialkyl phosphines, trialkyl phosphines, cycloaliphatic phosphines, aromatic phosphines, and phosphorous-containing alicyclic compounds.

Application No. 10/741,324
Amendment dated: November 29, 2006
Reply to Office Action of August 29, 2006

129420-2

37. (Currently Amended) A method for the continuous preparation of 4,4'-diisopropylbiphenyl, said method comprising:

(a) preparing a dealuminated acidic zeolite catalyst having a molar ratio of SiO₂ to Al₂O₃ in a range between about 20 to 1 and about 500 to 1, and determining an acidic site distribution for said catalyst, said distribution comprising 35% or more sites having an activation energy of ammonia desorption in a range between about 145 kJ/mol and about 170 kJ/mol;

[[(a)]] (b) continuously introducing into a flow reactor containing a solid bed of said [an acidic] dealuminated acidic zeolite catalyst having a ratio of SiO₂ to Al₂O₃ in a range between about 20 to 1 and about 500 to 1, said catalyst comprising acidic sites, at least 35% of said sites having an activation energy of ammonia desorption in a range between about 145 kJ/mol and about 170 kJ/mol, at a temperature in a range between about 180°C and about 320°C and a pressure between about 2 and about 50 atmospheres, (i) a solution comprising biphenyl and at least one inert hydrocarbon solvent, (ii) propene, and (iii) an inert diluent gas, said solvent having a boiling point in a range between about 180°C and about 320°C, said solution being introduced into said flow reactor at a feed rate corresponding to about 0.1 to about 5 catalyst bed volumes per hour, said propene being introduced at a feed rate corresponding to between about 1 and about 10 moles of propene per mole of biphenyl; and

[[(b)]] (c) continuously removing from the flow reactor an effluent stream comprising product 4,4'-diisopropylbiphenyl, inert hydrocarbon solvent, and inert diluent gas.

38. (Original) The method according to claim 37 wherein said acidic zeolite catalyst is prepared by a method comprising a step in which an acidic zeolite is treated with ammonia prior to its use as an alkylation catalyst.

39. (Currently Amended) A method for the preparation of an acidic zeolite catalyst, said method comprising:

(a) dealuminating a precursor zeolite by steam treatment of the precursor zeolite followed by exposure of the steam treated catalyst to aqueous acid to afford an acidic zeolite having a ratio of SiO₂ to Al₂O₃ in a range between about 20 to 1 and about 500 to 1; and

(b) treating the acidic zeolite of step (a) with a volatile basic agent to afford an acidic zeolite catalyst:

Application No. 10/741,324
Amendment dated: November 29, 2006
Reply to Office Action of August 29, 2006

129420-2

(c) determining an acidic site distribution for said catalyst, said distribution comprising 35% or more sites having an activation energy of ammonia desorption in a range between about 145 kJ/mol and about 170 kJ/mol to afford an acidic zeolite catalyst comprising acidic sites, at least 35% of said sites having an activation energy of ammonia desorption in a range between about 145 kJ/mol and about 170 kJ/mol.